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TWELFTH QUARTERLY TECHNICAL PROGRESS REPORT

ON

THE NUCLEAR PROPERTIES OF RHENIUM

FOR

DEPARTMENT OF THE NAVY
BUREAU OF WEAPONS
WASHINGTON 25, D. C.

under

CONTRACT NO.: NOas 60-6021-c

for the period

September 8, 1962 through December 8, 1962

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ABSTRACT

A complete decay scheme of Au^{196} and Au^{196*} is proposed. This decay scheme is based on shell model calculations and experimental measurements of the emitted gamma rays from Au^{196} and Au^{196*}. The half-life of Au^{196*} was found to be 10.4 hours. The absolute intensity of each gamma ray associated with the decay of Au^{196} and Au^{196*} was obtained.

INTRODUCTION

Since gold is used as a standard in many neutron cross section measurements, a knowledge of the complete decay scheme of Au^{196} and Au^{196*} is helpful in the determination of (n, 2n) cross sections. During this quarter the gamma rays associated with the decay of Au^{196} and Au^{196*} were studied. A complete decay scheme is proposed.

EXPERIMENTAL WORK

A 97.2 milligram circular gold foil, was placed against the tritium target of the Texas Nuclear 14.1 Mev neutron generator (1). The foil was irradiated for a period of two hours at a beam current of 250 microamps. The pulse-height spectrum of the gamma rays from the decay of Au^{196} and Au^{196*} was obtained with a 1 3/4" diameter x 2" long NaI(Tl) crystal and a multichannel analyzer. Frequent measurements of the pulse-height spectra were continued for 12 days.

Since 14.1 Mev neutrons can induce the (n, p) reaction in gold, a chemical separation which removed Pt^{197} from the sample was performed. The procedure utilized ethyl acetate extraction of the 6N hydrochloric acid solution of the gold foil which gives essentially complete decontamination from

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* The asterisk refers to the excited state of Au^{196}.

platinum radioactivities (2).

The foil was dissolved in aqua regia, platinum carrier was added and the solution was evaporated to dryness. The residue was taken up in 6N hydrochloric acid, mixed with an equal volume of ethyl acetate and shaken in a separatory funnel. After separation from the initial aqueous layer, the gold-bearing organic phase was washed with two additional volumes of 6N hydrochloric acid solution. The residue of the evaporation of the ethyl acetate solution was taken up in dilute nitric acid, transferred to a counting container and evaporated to dryness. Recovery of the gold radioactivity was found to be complete. A source prepared from the acqueous layer, which contained the platinum carrier, was found to be inactive.

TREATMENT OF DATA

The pulse-height distributions that were obtained for the foil after isomeric transition activity had decayed out were corrected back to a time at which the gamma rays from the isomeric transition were present in the pulse-height distributions. The difference between these two curves gave the pulse-height distribution caused by gamma rays from the isomeric transition alone. A plot of the count rate per channel in each peak versus time demonstrated that all of the residual peaks had the same half-life.

The intensity of the highest energy gamma ray from the isomeric transition was determined from known values of the photofraction and the crystal efficiency. The intensities of the lower energy gamma rays were determined in a similar manner following the subtraction of pulses contributed to each photopeak from higher energy gamma rays.

RESULTS

Figure 1 shows the pulse-height spectrum of the gold sample obtained immediately after irradiation. Figure 2 shows the pulse-height spectrum of the same sample taken six days after irradiation. It is seen in Figure 2 that the gamma rays emitted from the isomeric transition of Au^{196*} have decayed out. After subtracting the background from each channel, the count rate per channel in Figure 2 was multiplied by 2.16 ($e^{\lambda t} = 2.16$, where $\lambda = 0.131\ day^{-1}$ is based on $T_{1/2} = 5.3$ days for Au^{196}, and where t is the time difference between the pulse-height spectrum shown in Figure 1 and that in Figure 2.) The background was added to the pulse-height spectrum that was obtained and the resulting spectrum was subtracted from the spectrum shown in Figure 1. The difference between these two spectra is shown in Figure 3. This difference is the pulse-height spectrum of the gamma rays emitted from the isomeric transition of Au^{196*}. It is seen that there are six gamma rays (38 $\pm$ 6 Kev, 77 $\pm$ 6 Kev, 155 $\pm$ 6 Kev,

193 ± 6 Kev, 232 ± 6 Kev, 270 ± 6 Kev) associated with the isomeric transition. The uncertainty in the energy of each gamma ray line depends on the linearity of the multi-channel analyzer. A test of the analyzer showed that the instrument was linear with an uncertainty in energy of one channel width.

The method for the determination of the half-life of the isomeric transition was outlined in a previous section. Figure 4 shows a plot of the count rate at each peak versus time. An average value of the least squares fit for the half-life of all the gamma rays was found to be 10.4 hrs. This value for the half-life is slightly lower than that reported by other workers (3).

The method for the determination of the absolute number of the photons of each energy was also described previously. Table I shows the intensity of each gamma ray. The limit of the uncertainty in the absolute number of the photons of each energy is ± 5%. The primary source of error was in the estimation of the contributions from higher energy photons to the lower energy regions in the pulse-height spectrum.

Based on the number of gamma photons shown in Table I, a decay scheme for Au^{196} and Au^{196*} was constructed as shown in Figure 5.

TABLE I

Intensities of the Gamma Rays Associated With the Decay of Au^{196} and Au^{196*}

Energy, Kev	Photon Intensity Per Unit Time	Half-life
38 ± 6	55×10^3	10.4 hours
77 ± 6	525×10^3	10.4 hours
155 ± 6	340×10^3	10.4 hours
193 ± 6	274×10^3	10.4 hours
232 ± 6	88×10^3	10.4 hours
270 ± 6	73×10^3	10.4 hours
337 ± 6	354×10^3	5.3 days
356 ± 6	354×10^3	5.3 days
429 ± 6	270×10^3	5.3 days
514 ± 6	26×10^3	5.3 days
693 ± 6	125×10^3	5.3 days

DISCUSSION OF RESULTS

In a previous report (4), some possible exicited states of Au^{196} were calculated by the Blatt and Weisskoff formula (5) for isomeric transition. This formula, however, according to M. Goldhaber and R. D. Hill (6) is incomplete. It was used, however, as a guide for the assignment of the various states of Au^{196}. For example, by using the formula to calculate the energy difference between the h 9/2 and P 1/2 states for a neutron electric - $(2)^4$ - pole transition, it was found that the energy difference was 250 Kev as compared to 270 Kev observed experimentally. The calculated energy difference between the f 7/2 and P 1/2 states for M_3 neutron transition was 52 Kev; the observed energy difference was 77 Kev. The calculated energy difference between the h 9/2 and d 5/2 states for E_2 proton transition was 33 Kev and the experimentally determined value was 38 Kev.

Since Au^{196} is an odd-odd nuclei there remains a question whether the ground state is d 3/2 or P 1/2. Gold-197 has an odd proton and therefore its ground state is d 3/2. Furthermore, the first excited state of Au^{197} is known to be 77 Kev. (7). On the other hand, Hg^{197} has an odd neutron and therefore its ground state is P 1/2 (6). Thus, based on Blatt and Weiskoff's formula, it appears that the ground state of Au^{196} should be

P 1/2, whereas experimentally, as in the case of Au^{197}, the ground state should be d 3/2.

It is seen from Table I that the sum of the 155 and 232 Kev photons is different from the number of 38 Kev photons which populate the 232 Kev level. An internal conversion coefficient of 6.7 accounts for this difference. This value of the internal conversion coefficient compares very well with that calculated by M. E. Rose (8) for E_2 transition. The internal conversion coefficient as interpolated from Rose's tabulations was 7.0. It is also seen from Table I that the number of 77 Kev photons is not equal to the sum of the 193 and 155 Kev photons. An internal conversion coefficient for either E_2 or M_3 transitions would not account for this difference. Experimentally the conversion coefficient should be

$$\alpha_T = \frac{N_{155} + N_{193}}{N_{77}} = \frac{340 \times 10^3 + 274 \times 10^3}{525 \times 10^3} \qquad (1)$$

$$= 1.17 \ ,$$

where α_T is the total internal conversion coefficient. If α_T is assumed to be composed of contributions from E_2 and M_3 transitions, then one can write

$$\alpha_T = a_1 \alpha(E_2) + a_2 \alpha(M_3) \qquad (2)$$

where a_1 and a_2 are the relative intensities of the 77 Kev photons emitted by E_2 and M_3 transitions. By using values for $\alpha(E_2)$ and $\alpha(M_3)$ from Rose's tabulations (8), it is found that $a_1 = .413$ and $a_2 = 0.587$. Thus, the ratio $M_3/E_2 = 1.42$.

It has been reported (3) that there was no positron emission in the decay of Au^{196}. On a theoretical basis, the difference between the mass number of Au^{196} and that of Pt^{196} is 0.00 122 AMU (9) or 1.138 Mev which makes positron emission possible. Figure 1 shows that 0.514 Mev gamma rays were detected. Since each position results in two 0.511 Mev gamma rays, it is seen that the positron emission in the decay of Au^{196} is 1.7% of the total modes of decay. It is not known whether or not the positron carries away all the excitation energy. It was also found that Au^{196} decays by beta emission 35.3% of the time and by electron capture 63% of the time. The values reported by J. M. Hollander et al. (3) for these modes of decay are 20% and 80%, respectively.

Figure 1 also shows a peak at 0.693 Mev. This indicates that after the Au^{196} nucleus captures an electron, the resulting Pt^{196} atom de-excites by the emission of a photon of 0.693 Mev

as well as by the emission of two photons (0.356 Mev and 0.337 Mev) in cascade. The decay scheme presented in the compilations of J. M. Hollander, et al. shows these two photons in cascade but does not show the 0.693 Mev photon.

During the course of this study, it was shown that only gold activity was produced in appreciable amounts by the interaction of 14 Mev neutrons with gold. It was also shown that the gamma rays observed during the decay of this induced radioactivity has the correct half-lives for Au^{196} and Au^{196*} and fitted the theoretically derived level schemes for these isotopes. Together, these data demonstrate the fact that gold is a good standard for the determination of (n, 2n) cross sections.

FUTURE WORK

The large NaI(Tl) crystal was received recently and the capture gamma ray work is being undertaken.

PERSONNEL

Due to the graduation of R. A. Karam, this work will be continued under the direction of W. H. Ellis.

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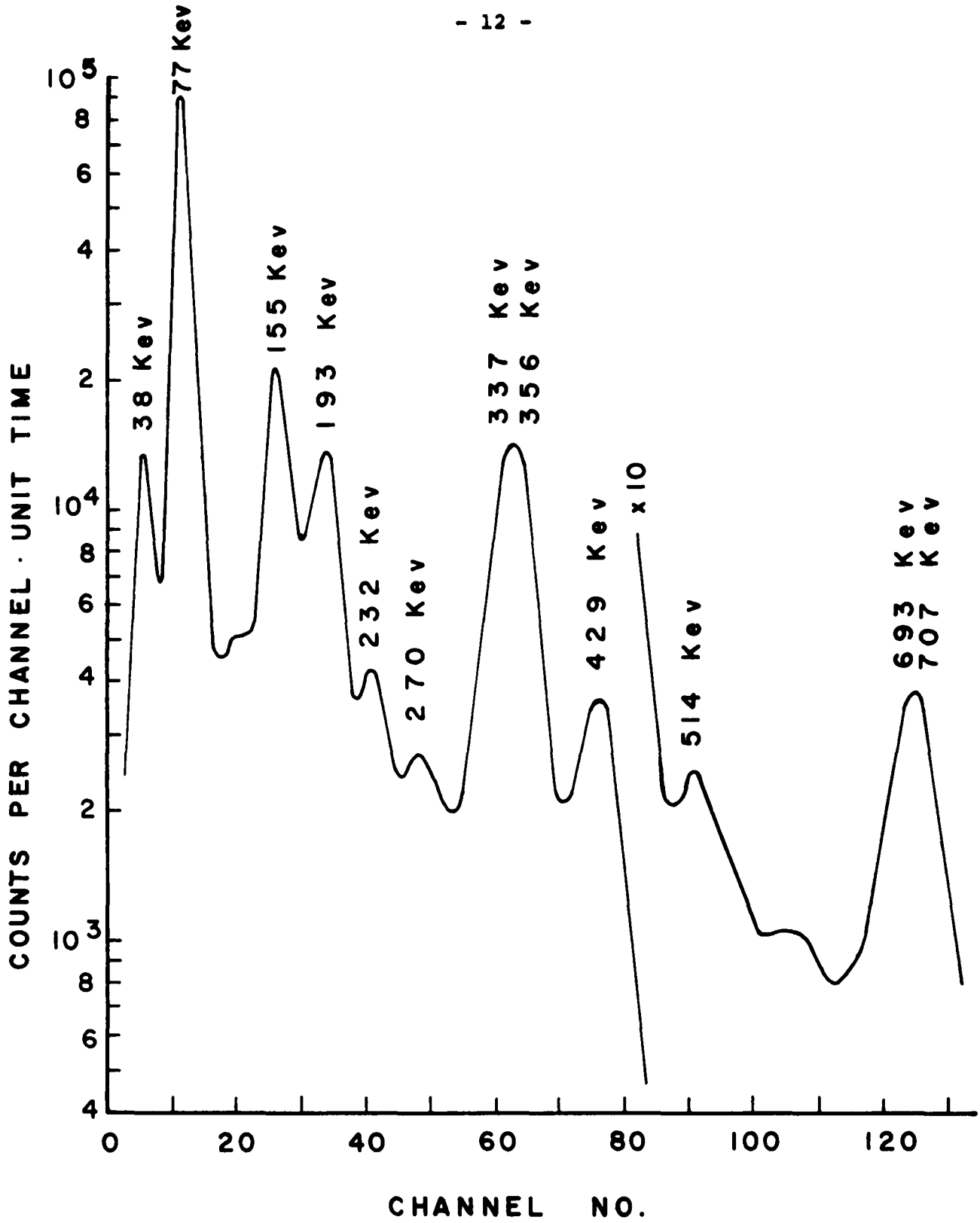

FIGURE 1. PULSE-HEIGHT SPECTRUM OF Au^{196} AND Au^{196*}

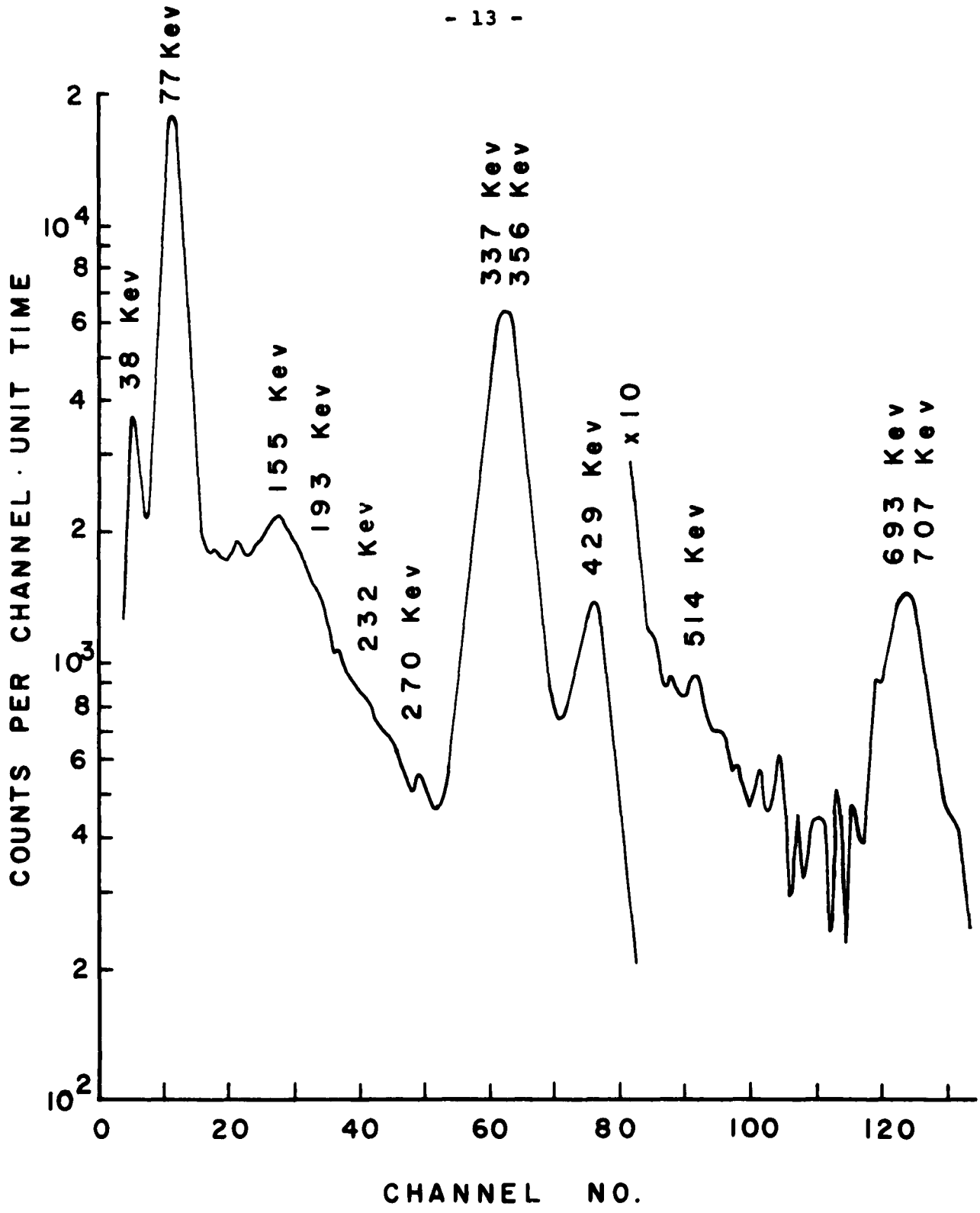

FIGURE 2. PULSE-HEIGHT SPECTRUM OF Au^{196}

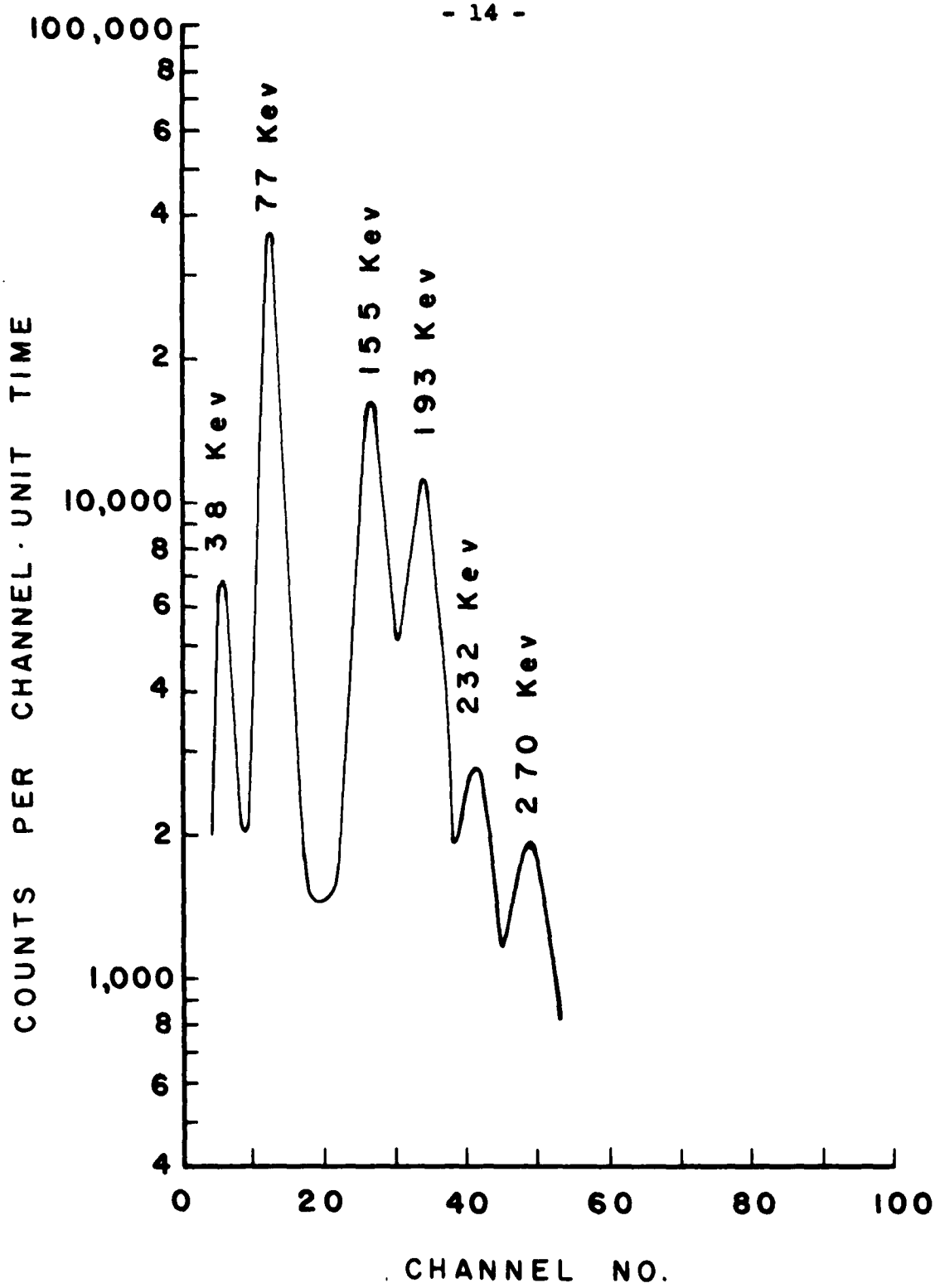

FIGURE 3. PULSE-HEIGHT SPECTRUM OF Au^{196*}

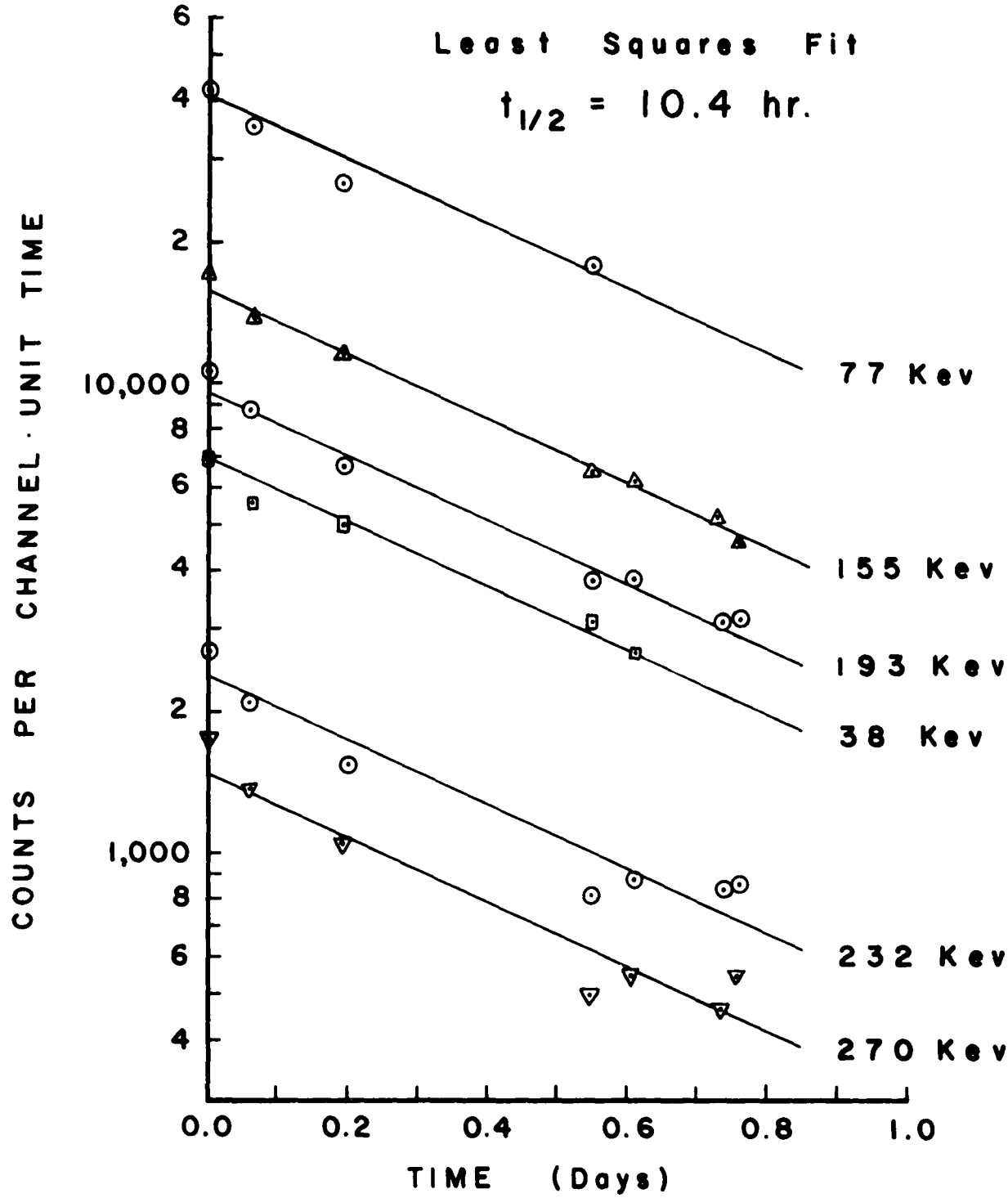

FIGURE 4. DECAY OF GAMMA RAYS FROM ISOMERIC TRANSITION OF AU^{196} VS. TIME

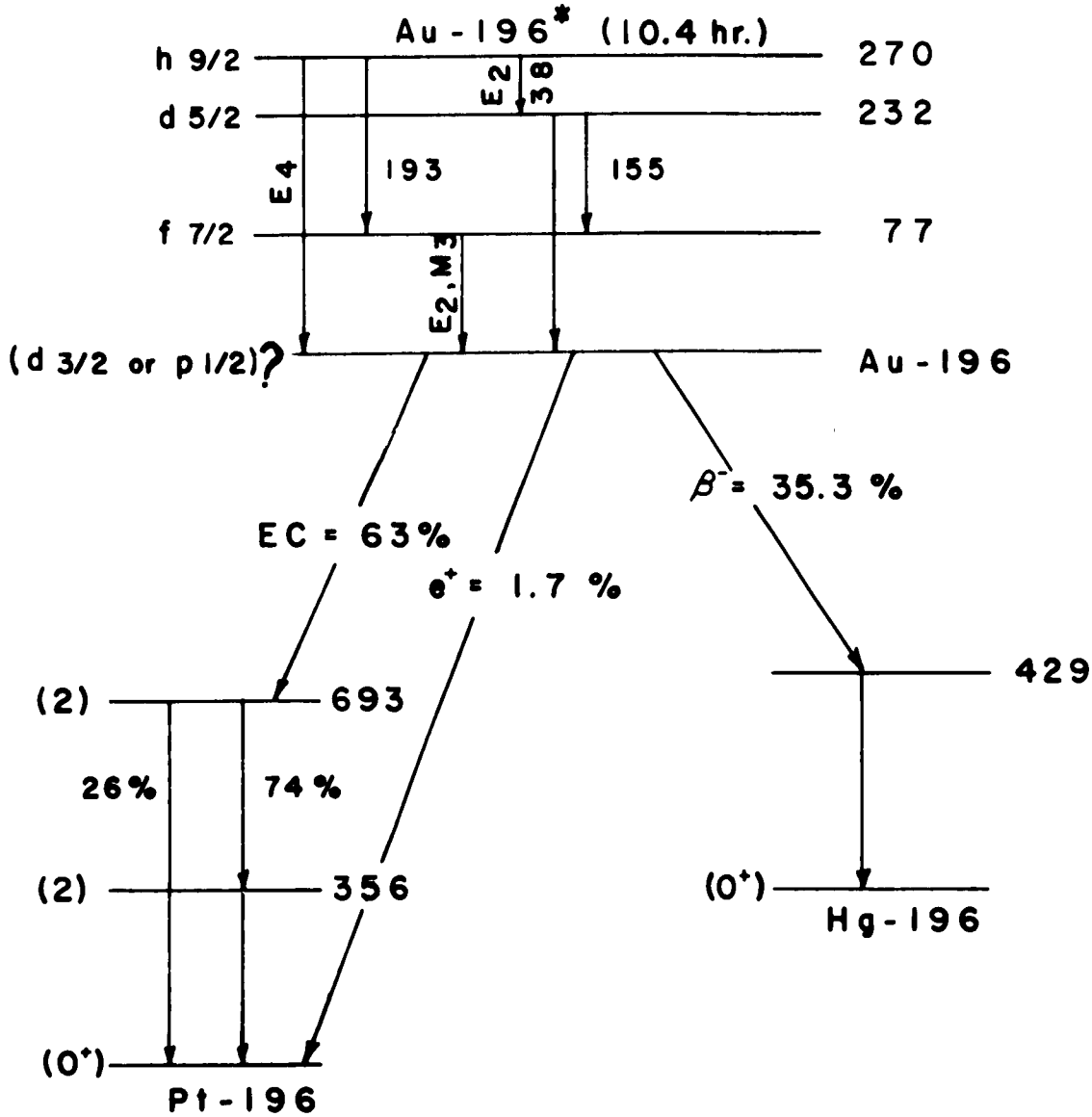

FIGURE 5. DECAY SCHEME OF AU^{196} AND AU^{196*}